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**"PASSIVE VIBRATION DAMPING MATERIALS: PIEZOELECTRIC CERAMIC
COMPOSITES FOR VIBRATION DAMPING APPLICATION"**

GRANT NO. N00014-92-J-1391

Annual Technical Report

Period : January 1, 1992 to January 31, 1993

TO:

DEPARTMENT OF THE NAVY
OFFICE IF NAVAL RESEARCH

FROM:

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Materials Research Laboratory
The Pennsylvania State University

February 1993

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ABSTRACT

For application of the shunted passive piezoelectric damping of structural composites, piezoelectric ceramic fibers (PZT and its family) of several tens of micron diameter were made using sol-gel spinning process. Both acetate and acetylacetonate processes were examined as lead sources. Using the properly chosen spinneret and fiber drawing apparatus, PZT and modified PZT fibers of 10 to 100 μ m diameter have been successfully produced. Different heat treatments gave different microstructure. Modeling analysis was also performed to estimate achievable damping levels for the composites. The results indicates high damping potential for shunted piezoelectric passive damping composites.

1. INTRODUCTION

This project was initiated in January 1990 with the purpose of exploring new methods of vibration absorption that may be effected by using a composite system with a ferroic solid as the active phase. The choice of ferroic solid controls the major damping mechanisms, whereas the matrix material provides mechanical strength as well as stress transfer to the active phase.

Our choice for the active phase was a piezoelectric ceramic, lead zirconate titanate (PZT). The principle of shunted passive piezoelectric damping was demonstrated using PZT ceramics in both theory and experiment during the first and second years of the contract.(1,2) In passive energy dissipation applications the electrodes of the piezoelectric ceramics are shunted with some electric conductance, hence the term shunted piezoelectric. The second year's effort was directed at studying the way in which PZT fibers could be incorporated into structural materials to achieve shunted piezoelectric damping. Fine PZT tubes (1.2mm diameter) which were available on the market were obtained to demonstrate PZT incorporation in structural materials. Two tube module in polymer matrix with external resistance was made first, and the attempt was made to produce PZT tubes in glass fiber reinforced epoxy. One of the methods most practical for large-scale production with structural polymeric materials is to use piezoelectric ceramics in fiber form, which must have high electro-mechanical coupling, whose diameter is comparable with reinforcing fiber materials used in the structural composites. The demonstration of sol-gel derived PbTiO_3 and PZT fiber (3,4) indicated that the sol-gel route was the optimum way to produce sub-100 μ m diameter piezoelectric ceramic fibers.

The third year's (this report period) effort, therefore, was aimed at producing fine PZT fibers by the sol-gel method and its process optimization, at establishing methods to pole and to characterize such fine fibers with diameters less than 100 μ m.



February 17, 1993

Dr. Lawrence Kabacoff
Scientific Officer Code: 1131M
Office of Naval Research
800 North Quincy Street
Arlington, VA 22217-5000

Dear Dr. Kabacoff:

Enclosed are three copies of the Final Technical Report of Grant No. N00014-92-J-1391.

Thank you for your support of the project. Please let me know if you have any questions.

Sincerely,

Shoko Yoshikawa
Senior Research Assistant

SY:jm

Enclosures

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A portion of the effort was also aimed at establishing modeling methods to estimate achievable damping levels, the shape of frequency-dependence, and the effects of complex stress states and shunting network topology.

2. ACHIEVEMENTS FOR THE PERIOD FROM JAN.1,1992 TO JAN.31,1993

The achievements in this period were multifaceted. First, sol-gel derived PZT and PZT family fibers formation processes were studied and characterizations were performed. Second, a poling method for fine continuous fiber was explored using extruded PZT-5H fiber with a diameter of $120\mu\text{m}$. Last, lamina modeling with various loading of PZT fibers were performed using a longitudinally poled fiber model. An alternate method, "coating" structural fibers with piezoelectric material, was also examined.

2-1. Sol-gel Derived PZT Family Fibers

Recognition of the need to produce micro-scale active components has been increasing recently with increased interest in smart materials and adaptive structures. State-of-the-art ceramic extrusion technique now offers $100\mu\text{m}$ diameter PZT semi-continuous fibers(5). We have chosen the sol-gel route to produce PZT fiber of less than $30\mu\text{m}$ diameter by spin-drawing PZT solutions at proper viscosity.

The first series of experiments was performed using lead acetate trihydrate as a lead source (designated "acetate" process). In $\text{Pb}(\text{Zr}_x\text{Ti}_{1-x})\text{O}_3$, compositions close to the morphotropic phase boundary exhibit high dielectric constant and high electromechanical coupling coefficients. In addition to the fiber formation of $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3$, two other formulations with small addition of niobium(Nb) or cadmium borate were explored to increase dielectric constant and electromechanical coupling by controlling grain growth and grain boundary conditions.

PZT precursor solutions in the form of viscous resins were extruded through a spinneret with 12 holes $200\mu\text{m}$ in diameter under pressure of approximately 100 psi. Continuous amorphous PZT fibers obtained were in the range of 5 to $100\mu\text{m}$ diameter. Several firing conditions were examined to study crystallization and grain growth. Pure perovskite phase was obtained by rapid thermal annealing at 700°C , though conventional (longer) heat treatments were necessary to promote grain growth of the fibers. Detailed information of this series of experiments is summarized in Appendix A.

The second series of experiments was aimed at producing larger quantities of fiber and the evaluation of new a lead source. Although acetate process has been studied since the

early eighties for thin film PZT formation, the number of preparation steps and total preparation time was long due largely to the multiple distillations which are necessary to remove the esters formed from the lead and PZT precursor solutions. Selvaraj, et. al. at Penn State University's Material Research Lab have shown that use of modified metal chelate complexes, such as lead acetylacetonate complex, instead of lead acetate trihydrate gave more stable PZT precursor solution in addition to a dramatic reduction of reflux steps(6). Therefore, we have made PZT and Nb-PZT solutions with both acetate and acetylacetonate (acac) processes for comparison. Although the difficulty in controlling the viscosity of the precursor resins still existed at the fiber formation stage, a 100 μ m spinnaret diameter produced continuous fibers with an average diameter of 30 μ m. Amorphous fibers were fired at various schedules, at the same time pellets were made from powders prepared from the same precursor solutions for comparison.

There was some difficulty in performing dielectric measurements on such fine fibers, because the fiber geometry gave rise to very small capacitances (order of 10^{-16} F) and to unusual fringing effects in the electrical field. We have developed modified parallel plate fixture to perform single fiber dielectric measurements. A paper describing detailed experimental results and discussing the second series of experiments is in preparation.

2-2. Poling method

Once high quality semi-continuous PZT fibers are formed, a continuous poling is necessary to produce longitudinally poled fibers. Serial poling experiments were performed using conventional mixed oxide PZT extruded fiber of 120 μ m diameter (obtained from CPS). In the serial poling process, the single fiber is considered to be composed of several sections of shorter length, and the whole fiber is poled by poling one section after another in a sample holder specially designed for poling. The holder contains two soft conductive rubber rings which are used as electrodes. The separation of the two rings can be adjusted according to the applied field.

Resonance measurements were carried out to determine the degree of poling. A center segment (5mm length) of a fiber (approximately 3cm) was excited to observe a fundamental resonance peak. In this method the measured capacitance of the fiber is ten times its actual capacitance, and the measured resonance peaks included the values of the clear overtone (See Figure 1 and 2). To obtain piezoelectric properties, the measured parameters must be corrected for the partial fiber excitation and the holder's capacitance. The modification to obtain an effective k_{33} from the partial excitation method is shown below:

$$\frac{k_{33}^2}{1-k_{33}^2} \cdot \frac{1}{L} = \frac{k_{pe}^2}{1-k_{pe}^2}$$

where, k_{33} is coupling coefficient of the fiber, k_{pe} is the coupling coefficient of the partially excited fiber, l is the length of excited section and L is the total length of the fiber. The measured properties of the extruded PZT-5H fiber poled at 2kV/mm for 10 minutes are as follows: $K_{33}'=2200$, $\tan\delta=0.024$, $v_{33}^D=3800$ m/s, $Q_m=35$, $k_{33}=0.68$, $d_{33}'=350$ pC/N (K_{33}' and d_{33}' are the effective values).

This poling method is feasible for fibers with sufficient strength. Since sol-gel derived fibers are not at the stage that they can be handled with ease, the poling study has been carried out using larger diameter straight fibers.

3. MODELING

A two-part approach has been followed in analytical modeling for the prediction of damping of laminates with embedded shunted piezoelectric fibers. The first part involves micromechanical modeling to predict the stiffness and damping properties of the composite (lamina) from the constituent properties. The composite is a three-phase material, composed of the reinforcing fibers (glass), epoxy matrix and piezoelectric fibers. Several micromechanical models predicting lamina stiffness properties from the constituent properties for two-phase composites are available in the literature (7,8). The three-phase composite is analytically treated as a two-phase material by considering the reinforced fibers (glass) and matrix as one phase (effective matrix) and the piezoelectric fibers as the other phase. In the case of glass fibers coated with piezoelectric ceramic, an effective fiber is formulated and treated as one phase with the matrix as the other phase. A dynamic micromechanical model based on transversely isotropic fibers and matrix is used twice in sequence, first to calculate the properties of the effective matrix or the effective fiber, and then to calculate the properties of the composite. The second part involves predicting the dynamics of the laminate based on the geometry, orientation, stiffness and damping properties of the lamina.

Appendix B gives a summary of the study with axially poled PZT fiber incorporated in a glass fiber reinforced epoxy matrix.

The possibility of sol-gel coating of PZT on optical fibers was shown by M.Sayer (9). The modeling effort thus included the following two cases as shown in Figure 3. The coating thicknesses were assumed to be 1, 5, and 10 μ m, while maintaining the same volume of epoxy. Table 1 shows three different coating thicknesses (thus different volume fractions of fiber) under the condition shown in Figure 3 (a), and one case which corresponds to

the Figure 3,(b) with the use of k_{31} as a comparison. In the case of PZT coating, however, it is important to mention that a $10\mu\text{m}$ coating of PZT gives an effective PZT volume fraction of over 50%, with a total fiber diameter of $30\mu\text{m}$. This high percentage of PZT will increase the density of the composite and reduce the effective composite moduli, as the volume fraction of the reinforcing glass fiber is only 6.7%. Figure 4 summarizes the strain energy distribution for the first five modes of a square cantilevered unidirectional laminate of fibers coated with $5\mu\text{m}$ of PZT. Please note that all the analyses were performed assuming the maximum electromechanical coupling of PZT and given an optimum external resistance.

4. CONTINUING AND FUTURE WORK

Effort is currently focused on the following areas:

1. PZT fiber process optimization and further characterization, particularly mechanical properties.
2. The feasibility of PZT coating on reinforcing fibers.
3. Materials and methods for electrical impedance matching.
4. Refinement of the modeling to explicitly include frequency dependence and complex strain effects in the composite.

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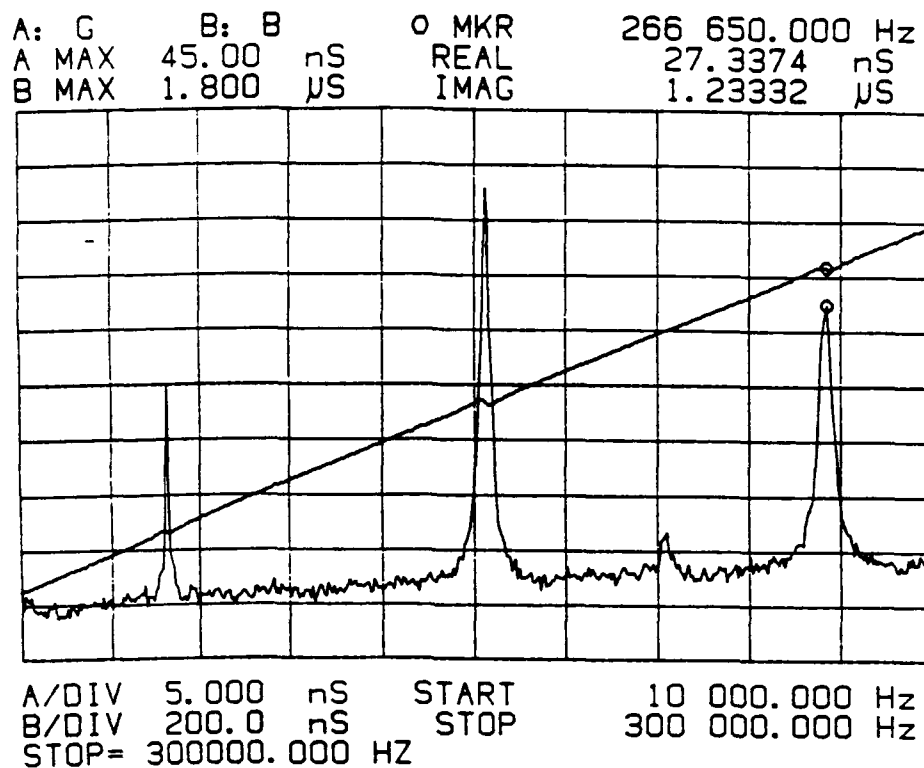


Figure 1. The resonance peaks (from the left, 1st, 3nd, 4th and 5th) of the PZT poled fiber.

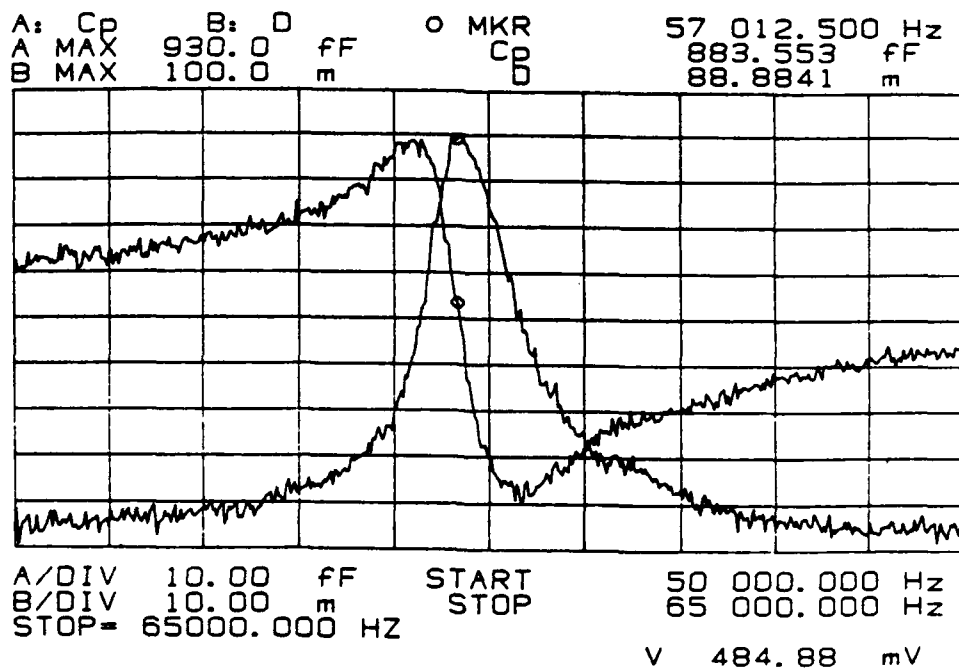


Figure 2. The capacitance and loss factor near the fundamental resonance frequency of the PZT poled fiber.

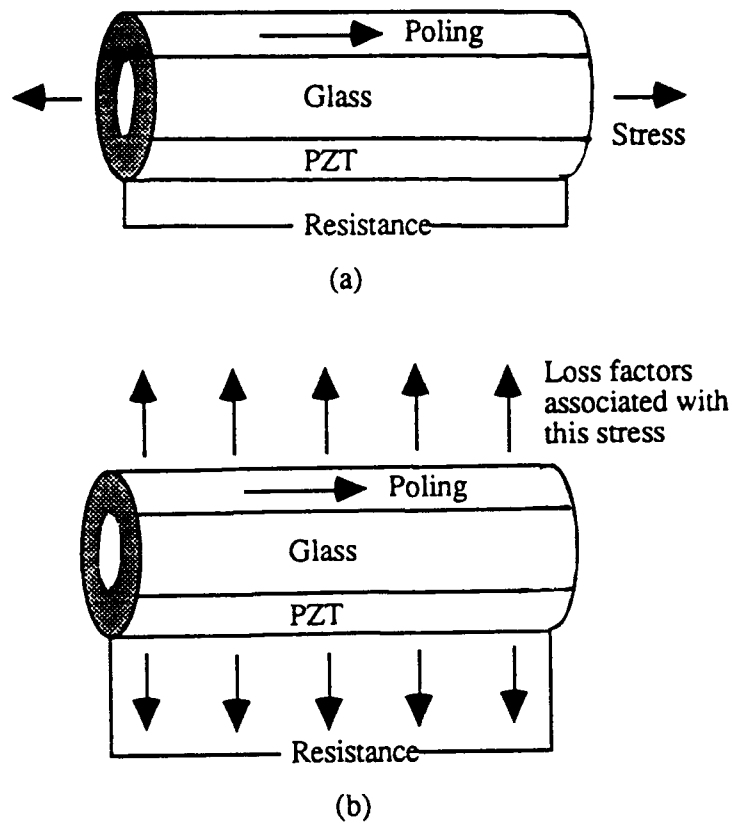


Figure 3. (a) Geometry, stress and poling directions for case 1, 2, and 3.
(b) Geometry, stress and poling directions for case 4.

Table 1. Compositions of composites used for the analysis

	PZT	S-glass	Epoxy	Coating thickness	Coupling coefficient used
case 1	18.33%	41.67%	40%	1 μ m	k33 only
case 2	45.0	15.0	40	5	k33 only
case 3	53.33	6.67	40	10	k33 only
case 4	53.33	6.67	40	10	k31 only

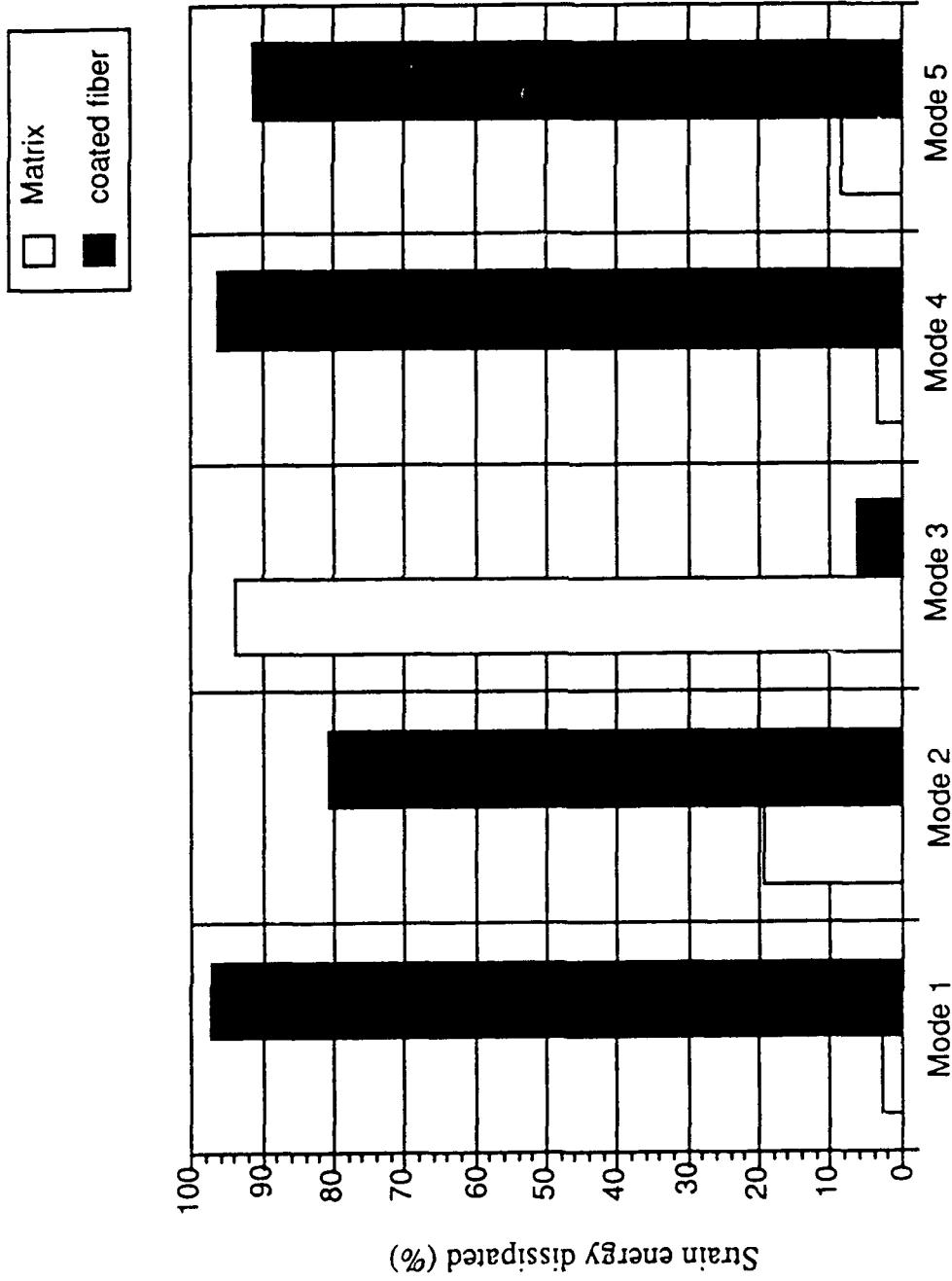


Figure 4 Strain Energy distribution for the first five modes for a square cantilevered unidirectional laminate with PZT coated fibers (coating thickness = 5 microns)

PIEZOELECTRIC PZT TUBES AND FIBERS FOR PASSIVE VIBRATIONAL DAMPING

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Abstract: Passive vibrational damping was demonstrated in piezoelectric lead-zirconium titanate (PZT) tubes incorporated into a hard epoxy matrix. For a poled tube the coupling constant, k_{33} measured from the resonance frequency, was about 0.66. Sol-gel methods were used to fabricate piezoelectric fibers of $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3$, and niobium and CdBO_3 substituted PZT.

Continuous and fine gel fibers of about 5 to 100 μm in diameter were prepared by extruding and drawing spinnable-viscous resins through a spinneret. These fibers fired at 600°C for 1 h exhibited well-crystallized perovskite phases of PZT. Fibers fired between 700° and 1250°C were dense with varying grain sizes (0.1 to 5 μm). The dielectric constant of $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3$ fiber fired at 700°C for 1 h was about 800. These fibers will be incorporated into structural materials to obtain maximum damping properties.

1. Introduction

In recent years, there has been growing interest, among researchers in various fields, to minimize and control vibration in machines and devices. Vibration can be significantly reduced by increasing the damping of the dominant modes of vibrations through passive and active damping mechanisms. In active vibrational damping devices, the vibration is minimized by using a certain active elements such as actuators that alter the dynamic response of the structure. These devices, however, require the use of special hardware and real-time control-design algorithms for individual structural elements. The active vibrational damping system is therefore complicated and is still in its developmental stages. By contrast, in passive vibrational damping devices, vibrational energy is dissipated through the added external damping media such as isolation- or constrained-viscoelastic layers. These devices are simple and offer a reliable solution for vibrational suppression over a limited range of frequency. A more realistic future damping control device is likely to have a balanced combination of both active and passive systems.

For passive and active damping applications, piezoelectric ceramics with large electromechanical coupling coefficients (k_j) are potential candidates [1,2]. Based on theoretical and experimental studies [1,2], it has been established that piezoelectric ceramics can provide a large mechanical loss factor and thus can be effective in passive damping. Until now, however, it is not apparent that how and in what forms these piezoelectric ceramics have to be incorporated into structural materials in order to achieve maximum damping. Hence, our goal is to develop methods that allow an effective piezoelectric-passive damping in large structural materials.

The first section of the paper describes the incorporation of commercially available fine PZT tubes in a relatively hard polymer matrix. The PZT tube-polymer configuration (1-3 composite) is such that maximum electromechanical coupling and hence an effective damping is achieved for optimum external resistance. The second section deals with the sol-gel processing of continuous PZT and modified PZT fibers, which would be incorporated into the polymer matrix in the future.

2. PZT/Polymer Composites

Piezoelectric-ceramic tubes (PZT-5H) obtained from Morgan Matroc, Inc. were used to study the passive vibrational damping. The dimensions of these tubes were: 1.28 mm outer diameter, 0.81 mm inside diameter and 10 cm length. In order to obtain maximum k_{33} from these tubes, 10 circumferential silver/glass electrodes of 1 mm in width and 1 cm apart were applied to each tube. These tubes were then fired at 800°C for 30 min. After firing, they were individually poled with an applied field of $\sim 10 \text{ kV/cm}$ at 80°C for

10 min. The coupling constant, k_{33} , calculated from the resonance frequency, for a poled tube was 0.66.

Fine silver wires were attached to the silver electrodes using a silver epoxy and cured at room temperature. A two-tube module constructed according to the above procedure is shown in Figure 1. A hard epoxy was applied to the two-tube module, leaving the silver wires outside the module for connecting to the external resistor, and keeping the hollow parts of the tubes without filling with any epoxy. The epoxy was cured at room temperature for 24 h, and then heat-treated at -65°C for 45 min.

The damping measurement was performed by exciting the middle portion of the sample. The intensities of resonance peaks were measured, while varying external resistance. Figure 1 shows the damping circuit with external resistors. The variation of mechanical loss factor with external resistance for the two-tube module is shown in Figure 2. The mechanical loss tangent, calculated from the resonance frequency (3500 to 4000 Hz) arises from the bending vibration of the sample. From the peak of mechanical loss tangent, it was found that the maximum piezoelectric damping was achieved for an external resistance of $\sim 8 \text{ M}\Omega$.

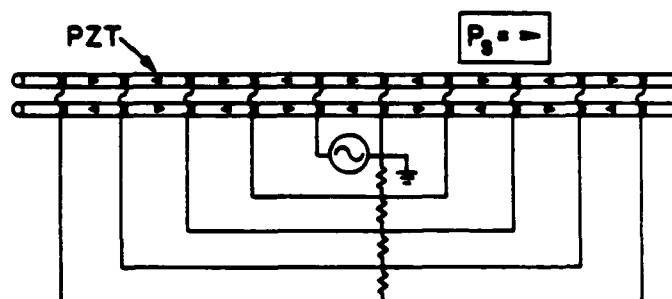


Figure 1 Schematic diagram of the poling configuration and resistive circuit for the two-tube module.

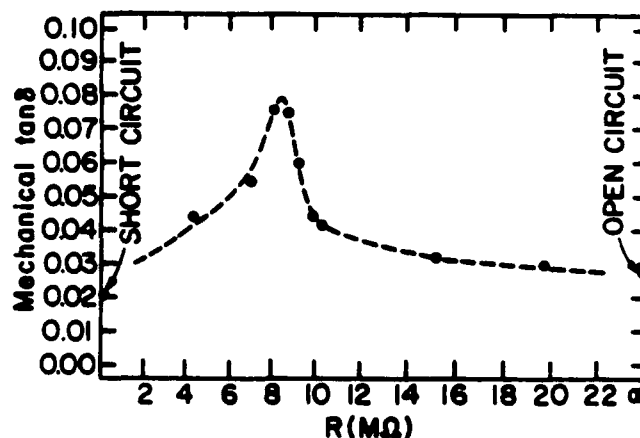


Figure 2 Mechanical loss tangent as a function of external resistance for the two-tube module.

3. Sol-Gel Fabrication of Continuous PZT and modified PZT Fibers

From practical viewpoint, incorporation of 1.28 mm diameter piezoelectric-ceramic tubes into structural materials leads to many problems of feasibility. Hence, efforts were directed towards producing fine piezoelectric-ceramic fibers, which can be easily embedded into various types of structural materials, like glass-fiber reinforced plastics.

In $\text{Pb}(\text{Zr}_x\text{Ti}_{1-x})\text{O}_3$, compositions close to the morphotropic-phase boundary ($x = 0.52$ to 0.55), exhibit high dielectric constants, and electromechanical coupling coefficients [3]. Addition of a few percentages of niobium enhances the dielectric and piezoelectric properties of PZT ceramics. Sol-gel processing of PZT fibers has gained much interest because of its simplicity, low processing temperature, chemical homogeneity and stoichiometry control and the ability to produce fibers of uniform microstructure [4,5]. Accurately-controlled microstructures and special shaping by chemical processes like sol-gel technique are essential for obtaining dense PZT ceramics for high-performance applications. The sol-gel ceramic materials of PZT and modified PZT in the fibrous form may exhibit increased response in small scale devices. Because of the volatility of lead at processing temperatures, PZT fibers cannot be fabricated by the melt process in the same way as silica fibers. PZT fibers have been prepared using a variety of precursors [6,7]. In this paper, we report the fabrication of continuous $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3$, and niobium and CdBO_3 substituted PZT [$\text{Pb}_{0.988}(\text{Zr}_{0.52}\text{Ti}_{0.48})_{0.976}\text{Nb}_{0.024}\text{O}_3$ and $97\% \text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3 + 3\% \text{CdBO}_3$] fibers. Different firing schedules were followed in order to obtain fibers with varying grain sizes.

Preparation of Viscous Resins for Fiber Drawing:

The scheme for the preparation of spinnable-viscous resins of PZT, and niobium and CdBO_3 substituted PZT is outlined in Figure 3. Lead acetate trihydrate [$\text{Pb}(\text{CH}_3\text{COO})_2 \cdot 3\text{H}_2\text{O}$], zirconium n-butoxide, $[\text{Zr}(\text{OBu})_4]$ 80% solution in 1-butanol, titanium isopropoxide [$\text{Ti}(\text{OPr}^i)_4$], niobium ethoxide [$\text{Nb}(\text{OC}_2\text{H}_5)_5$], cadmium acetate hydrate [$\text{Cd}(\text{CH}_3\text{COO})_2 \cdot x\text{H}_2\text{O}$] and boron methoxide [$\text{B}(\text{OCH}_3)_3$] obtained from Aldrich Chemical Company were used as the starting materials. Lead acetate trihydrate dissolved in 2-methoxyethanol was distilled off three times. A stoichiometric quantity of $\text{Zr}(\text{OBu})_4$ was added to the lead solution and refluxed at 125°C for ~6 h. $\text{Ti}(\text{OPr}^i)_4$, $\text{Nb}(\text{OC}_2\text{H}_5)_5$, $\text{Cd}(\text{CH}_3\text{COO})_2 \cdot x\text{H}_2\text{O}$ and $\text{B}(\text{OCH}_3)_3$ were then added to the Pb-Zr solution and again refluxed at 125°C for ~6 h to form the precursor solutions of PZT, and niobium and CdBO_3 substituted PZT. A solution of 1 ml of water and 1 ml of conc. HNO_3 diluted in 25 ml of 2-methoxyethanol was added to a vigorously stirred precursor solution of 0.2 M PZT or niobium or CdBO_3 substituted PZT. The solution was concentrated by stirring at $\sim 120^\circ\text{C}$, and then cooled to $\sim 40^\circ\text{C}$ to form a viscous resin. As a result of the foregoing treatment the viscous resin was suitable for extrusion and drawing. The viscosity of the resin in the fiber drawing region was at least 10^6 mPa.s.

Drawing and Final Consolidation:

The gel fibers were extruded through a spinneret (e.g., with 12 holes of 200 μm in diameter) at less than 100 PSI. The complete fiber drawing, fiber take-up spool and spinneret assemblies are illustrated elsewhere [8]. These fibers were stretched or drawn by mechanical means to less than 200 μm in diameter. The pulled gel fibers from the spinneret were collected on a rotating drum with a variable-speed control. The drawn gel fibers retained the shape of the spinneret because of the cohesive property of the resins. Because of the viscoelasticity of the resins, the drawn fibers were from approximately 1/20 to 1/3 of the spinneret nozzle diameter. The fibers obtained were of the order of 5 to 100 μm in diameter.

Rapid Thermal Processing:

Niobium substituted PZT fibers preheated at 400°C for 12 h were annealed by a rapid thermal process (RTP) in a Heat Pulse

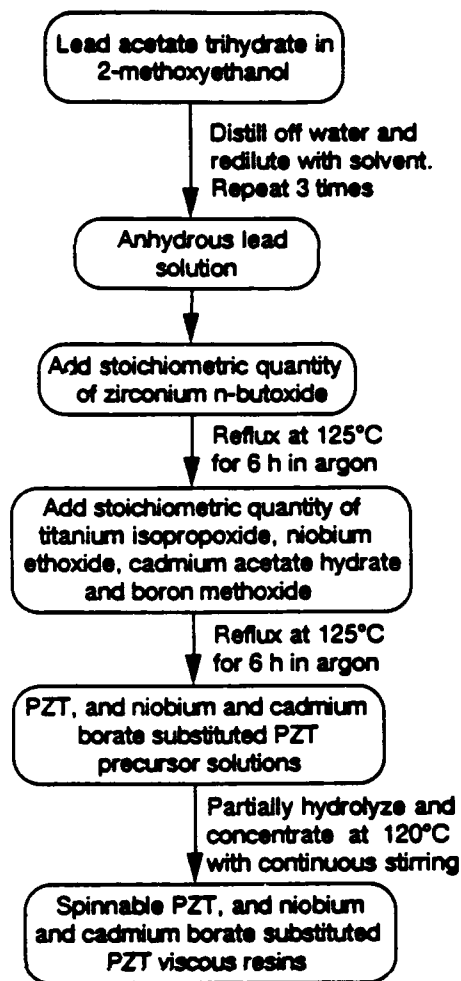


Figure 3 Scheme for the preparation of the spinnable PZT, and niobium and cadmium borate substituted PZT viscous resins.

210T rapid thermal annealer. These fibers were supported on a 4" Si wafer coated with Platinum. Before each run the system chamber was purged with high-purity oxygen. Typical run conditions were: i) a 10 s ramp to 700°C , ii) hold at 700°C for 10 s, and iii) cooled down to room temperature. No apparent reaction occurred between the fiber and the coated Si wafer during the brief RTP anneal.

Characterization:

Phase transformations and the weight loss of the gel fibers of PZT, and niobium and CdBO_3 substituted PZT obtained from the spinnable resins were studied using Perkin-Elmer differential thermal (Model DTA 1700) and thermogravimetric (Delta Series TGA7) analyzers interfaced with a computerized data acquisition and manipulation system. Phases crystallizing in the heat-treated samples were identified using a Scintag (Model DMC 105) diffractometer with Ni filtered $\text{CuK}\alpha$ radiation. The microstructure and the diameter of the heat-treated fibers were studied by a scanning electron microscope (ISI-DS 130, Akashi Beam Technology Corporation, Japan). The dielectric constant of single fiber of PZT was obtained using a precision capacitance bridge (Model GR 1621, General Radio, MA). Capacitance was measured using a three-terminal shielded measurement at 1 kHz. The test fixture capacitance was compensated by open-circuit subtraction. Fibers of 1 to 2 mm in length were attached to sputtered gold electrode pads using a silver paint.

The spun gel fibers were dried at room temperature and heated to 400°C at a heating rate of $1^\circ\text{C}/\text{min}$ to eliminate organic

constituents and most of the residual carbon. Figure 4 shows DTA and TGA curves for PZT fibrous gel previously heat treated at 400°C for 24 h. The pre-heated PZT gel exhibited a weight loss of ~3% in the temperature range of 50° to 700°C. The gel exhibited a sharp exotherm at 482°C, followed by a shoulder at 530°C in DTA. These peaks can be attributed to the crystallization of a pyrochlore phase and its conversion into a perovskite PZT. XRD results of the fibrous PZT gel heat treated at 500°C for different durations indicated the formation of pyrochlore and perovskite phases, while heat treatment at 600°C resulted in only the perovskite phase (Figure 5).

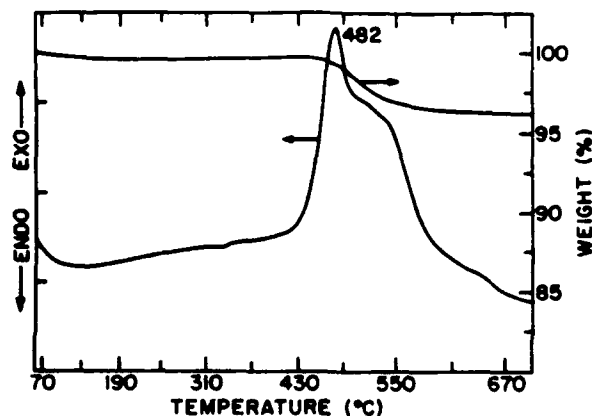


Figure 4 DTA and TGA curves for the fibrous PZT gel pre-heated to 300°C for 24 h.

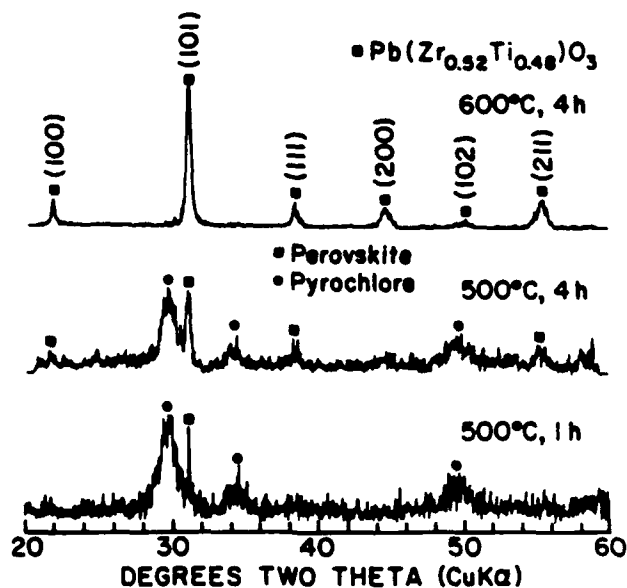


Figure 5 XRD patterns for the fibrous PZT gel heat-treated at different temperatures.

Scanning electron microscope (SEM) pictures of PZT, and niobium and CdBO₃ substituted PZT fibers heat treated at 700 °C for 1 h and etched with 1% HCl are shown in Figures 6(a & b), (c & d) and (e & f) respectively. Under higher magnification, PZT and niobium substituted PZT fibers showed fine grains of 0.10 to 0.2 μm (Figures 6(b) and 6(d)). Addition of CdBO₃ to PZT resulted

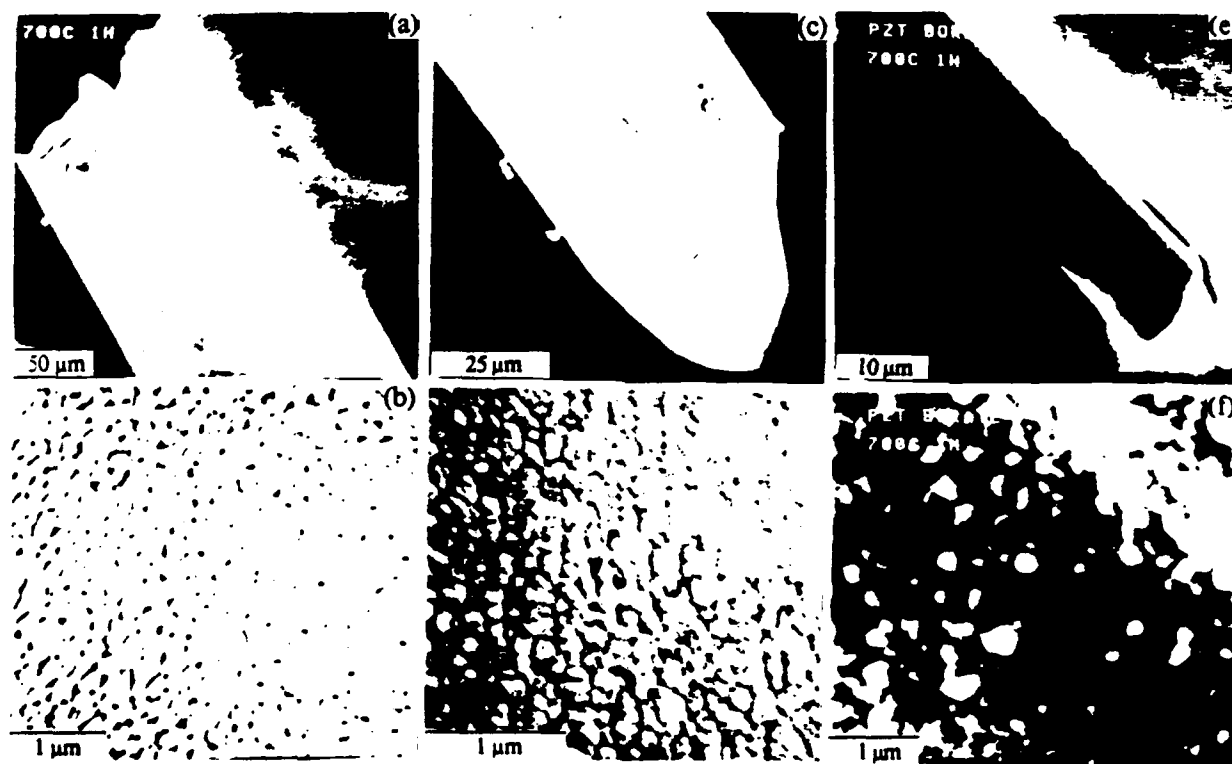


Figure 6 (a) & (b), (c) & (d) and (e) & (f) are respectively the low- and high-magnification micrographs of PZT, niobium and CdBO₃ substituted PZT fibers heat-treated at 700°C for 1 h.

in larger grains (~ 0.1 to $0.35 \mu\text{m}$) for fibers annealed at 700°C for 1 h (Figure 6(f)). Figure 7 shows the SEM micrographs of niobium substituted PZT fibers fired at 700°C for 10 s by the RTP ($\sim 4100^\circ\text{C}/\text{min}$). This resulted in a denser fiber with more uniform microstructure (grain size $\sim 0.1 \mu\text{m}$) compared to the one fired at a heating rate of $1^\circ\text{C}/\text{min}$ (Figures 6(b)). PZT fibers were also fired at 1250°C for 0.2 h in a closed lead oxide atmosphere. Lead zirconate powder was used as the source to create lead oxide atmosphere. SEM micrographs of these fibers (Figure 8) show that they are dense and possess grains of about 0.5 to $5 \mu\text{m}$.

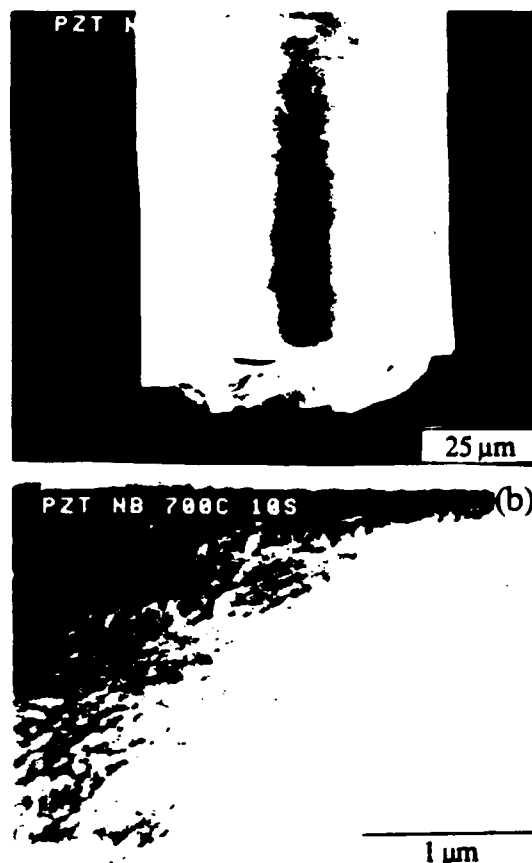


Figure 7 (a) & (b) are respectively the low- and high-magnification micrographs of the niobium substituted PZT fibers fast fired at 700°C for 10 s.

The room temperature dielectric constant of the PZT fiber fired at 700°C for 1 h was found to be ~ 800 . The corresponding loss value ($\tan\delta$) for the fiber was of the order of 0.06 to 0.08. These values are in agreement with those reported for sintered ceramics of the same composition [3]. Experiments are being carried out to pole these ultrafine PZT, and niobium substituted PZT fibers and embed them in polymer matrix for passive vibrational damping studies.

4. Conclusions

Piezoelectric-ceramic tubes, poled and embedded in a polymer matrix, exhibited passive damping with optimum external resistance. By sol-gel processes, uniform PZT, and niobium and CdBO_3 substituted PZT gel fibers of unlimited length were obtained from the spinnable-viscous resins by extrusion and drawing through a spinneret. The diameter of these fibers were between 5 and $100 \mu\text{m}$. The fibrous gels yielded well-crystallized PZT at 600°C . PZT, and niobium and CdBO_3 substituted PZT fibers fired at 700°C for 1 h were found to be dense with submicron (0.1 to $0.35 \mu\text{m}$) grains. PZT, and CdBO_3 substituted PZT fibers with grain sizes of

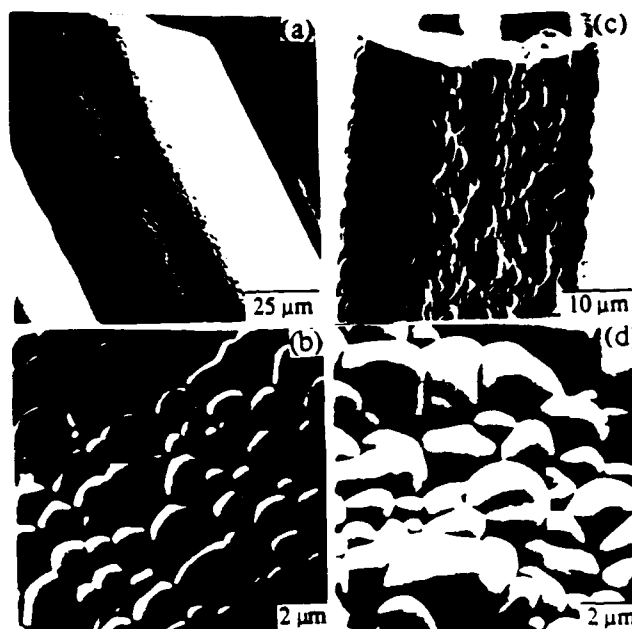


Figure 8 (a) & (b) and (c) & (d) are respectively the low- and high-magnification micrographs of PZT, and CdBO_3 substituted PZT fibers heat-treated at 1250°C for 0.2 h.

about 0.5 to $5 \mu\text{m}$ were obtained on firing the fibers in a closed lead oxide atmosphere at 1250°C for 0.2 h. The room temperature dielectric constant of the PZT fiber fired at 700°C for 1 h was about 800.

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APPENDIX B

Resistively-Shunted Piezoceramics for Passively-Damped Structural Composite Materials

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Abstract

The development of structural materials which also display high damping is an area of current research with potential for high payoff. Resistively-shunted piezoceramic fibers used as reinforcement in a structural composite material offer the potential to significantly increase vibration damping capability. In addition, the damping of such materials should be less sensitive to temperature than that of conventional non-structural damping treatments. This paper addresses the current status of an effort to develop such damped composite materials, including modeling aspects, performance limits, design guidelines, fabrication issues, and experimental results.

Initial design guidelines take the form of a modified modal strain energy method. With longitudinally-poled fibers, peak damping loss factors of 12% are theoretically attainable, even at low (30%) piezoceramic fiber volume fractions. Some piezoceramic fibers have been produced using a sol-gel method, while details of poling and shunting are under investigation. Initial data for flexure of small specimens show predictable damping levels and frequency dependence.

THE USE OF PIEZOELECTRIC MATERIALS with resistive shunting circuits to achieve passive vibration energy dissipation and resonant response reduction has been demonstrated by several researchers [1-4]. Resistively-shunted piezoceramics appear to offer several advantages over more conventional approaches to passive damping, including: 1) relative insensitivity to temperature; 2) tailorable frequency-dependence; and 3) high stiffness. A disadvantage includes the relatively high density of typical lead-based piezoceramics.

Because of high electroelastic coupling, the deformation of piezoelectric materials produces internal potential gradients. By placing electrodes on the material and shunting them through some finite resistance, current is allowed to flow, dissipating energy through joule heating.

When the dimensions of piezoelectric elements used for passive damping are comparable in magnitude to characteristic vibration lengths, element placement significantly affects achievable levels of structural damping. However, if the elements could be reduced in size and proliferated throughout a structure, possibly as reinforcement in a structural composite material, significant damping could be achieved with less sensitivity to placement.

This observation provided the motivation for the subject work, which addresses the development of resistively-shunted piezoelectric ceramic fibers as a means to increase the vibration damping properties of structural composite materials to significant levels. Fig. 1 shows a concept for such a fiber.

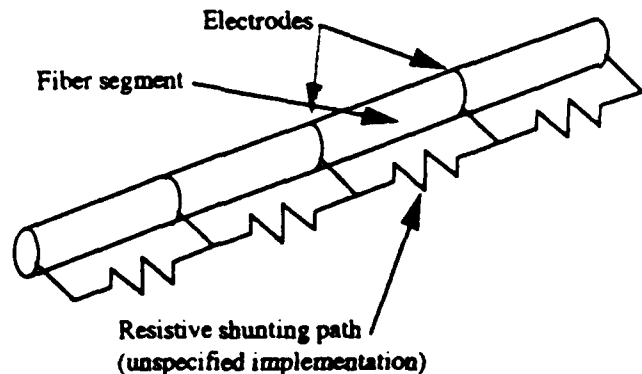


Figure 1: Concept for Resistively-Shunted Piezoelectric Fiber

Key challenges identified at the outset of this effort were associated with fabrication and modeling. Fabrication issues included: fiber production, poling and electroding; provision of an integral, tailorable resistive path; and integration into a composite material. Modeling issues included: estimation of achievable damping levels, shaping of frequency-dependence, and effects of complex stress states and shunting network topology. This paper focuses on the modeling issues.

One of the primary goals of the mathematical modeling portion of this effort was the establishment of achievable damping performance. If initial estimates, based on simplified models, were to indicate that the levels of damping possible were not of engineering significance, there would be little motivation to proceed. If, on the other hand, further investigation was warranted, more detailed models would be needed to design composite materials for specific applications, as well as to guide continuing materials development.

The initial modeling effort concentrated on the development of simple models that could be used to establish possible performance levels. This was done using a two-step process: the first was to determine effective loss factors for the piezoelectric fiber; the second was to use those loss factors in the estimation of modal damping for flexure of a composite panel. The following subsections discuss each of these steps in turn, as well as limitations of the models and future efforts.

Damping Performance Analysis: Effective Fiber Loss Factor

To a first approximation, the peak damping of a shunted piezoelectric fiber may be estimated by assuming that individual fibers in a composite experience a simple state of stress, namely pure longitudinal tension or compression, and that the stress is approximately uniform between two electrodes on the fiber. In this situation, the maximum damping achievable in the fiber alone may be treated as a material property—an effective longitudinal loss factor.

The damping of a composite material undergoing harmonic deformation may then be estimated as the sum of the damping in the constituent materials weighted by the relative contribution of each to total strain energy. Because the fiber modulus is typically much greater than that of the matrix material, most of the strain energy of deformation (often 80–95%) is found in the fiber. This is one of the primary motivations for seeking ways to increase the damping of reinforcing fibers.

As discussed in [4], interpretation of the operative physical dissipation mechanism as an anelastic relaxation permits the use of classical relations for analysis (such as those discussed in [5]) and the extension of established tools for design purposes. In this approach, the difference between the low-frequency modulus (E_r , relaxed, short-circuit) and the high frequency modulus (E_u , unrelaxed, open-circuit), is closely related to the peak material damping (η , loss factor) associated with that modulus. Eqs. 1a and 1b express both in terms of the relaxation strength, Δ :

$$E_u = E_r(1 + \Delta) \quad (1a)$$

$$\eta = \frac{\Delta}{2(1 + \Delta)^{1/2}} \quad (1b)$$

Note that the relaxation strength is closely related to the electromechanical coupling coefficient, k , as shown in Eq. 2:

$$\Delta = \frac{k^2}{(1 - k^2)} \quad (2)$$

The electroelastic relaxation strength corresponding to the longitudinal modulus may be found from consideration of the material constitutive equations, specialized to the case of a single non-zero (longitudinal) stress. The constitutive equations for a linear piezoelectric material relate the stress, T , and the electric displacement, D , to the strain, S , and the electric field, E , through several material properties. These properties include the elastic moduli, c^E , the piezoelectric constants, e , and the dielectric matrix, ϵ^S . Eq. 3 shows the general form of the constitutive equations:

$$\begin{aligned} T &= c^E - e^T E \\ D &= eS + \epsilon^S E \end{aligned} \quad (3)$$

Note that the equations are expressed in "vectorized" form (as opposed to tensorial). For poled piezoceramic materials, the "3" direction is taken by convention to be the direction of poling.

The material properties for PZT-5A, of particular interest for this project, are [6] (SI units):

$$c^E = 1.0e+11 \cdot \begin{bmatrix} 1.2100 & 0.7540 & 0.7520 & 0 & 0 & 0 \\ 0.7540 & 1.2100 & 0.7520 & 0 & 0 & 0 \\ 0.7520 & 0.7520 & 1.1100 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0.2110 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0.2110 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0.2260 \end{bmatrix}$$

$$e = \begin{bmatrix} 0 & 0 & 0 & 12.3000 & 12.3000 & 0 \\ 0 & 0 & 0 & 12.3000 & 12.3000 & 0 \\ -5.4000 & -5.4000 & 15.8000 & 0 & 0 & 0 \end{bmatrix}$$

$$\epsilon^S = 1.0e-08 \cdot \begin{bmatrix} 0.8107 & 0 & 0 \\ 0 & 0.8107 & 0 \\ 0 & 0 & 0.7345 \end{bmatrix}$$

The open-circuit moduli, c^D , may be found by taking $D=0$:

$$c^D = c^E + e'(\epsilon^S)^{-1}e \quad (4)$$

Similarly, the short-circuit modulus may be found by taking $E=0$. Note that, in practice, only the component of the electric field in the direction between electrodes is zero.

If the case of longitudinal fiber poling is considered, and the only non-zero stress is T_3 (in the direction of poling, or along the fiber axis), then the corresponding moduli are found to be:

$$\begin{aligned} \text{(relaxed, short circuit)} \quad E_r &= 5.34 \times 10^9 \text{ Pa} \\ \text{(unrelaxed, open circuit)} \quad E_u &= 10.54 \times 10^9 \text{ Pa} \end{aligned}$$

The corresponding relaxation strength and loss factor for the longitudinal modulus is then:

$$\begin{aligned} \text{(relaxation strength)} \quad \Delta &= 0.97 \\ \text{(longitudinal loss factor)} \quad \eta_L &= 0.35 \end{aligned}$$

Note that this approach yields a coupling coefficient, k_{33} , of 0.70, in agreement with the published value [6].

A similar approach may be used to address the case of radial poling and longitudinal deformation or, equivalently, longitudinal poling and transverse deformation, to yield a value for the transverse loss factor:

$$\text{(transverse loss factor)} \quad \eta_T = 0.06$$

Note that these values for η_L and η_T are ideal, peak loss factors attainable in practice over a limited frequency range. In composite design, other factors affecting the frequency-dependent behavior must also be addressed. These factors include resistive shunting, non-uniform strain over finite fiber segment lengths, and shunting network topology. The following subsections briefly address the general effects of each of these factors.

Resistive Shunting. The dynamics of the electrical RC shunting network result in frequency-dependent behavior. However, the value of the shunt resistance(s) may be specified by a designer to tailor or "tune" the frequency-dependence of damping to the application. Using the complex modulus representation of material properties ($E = E' + iE''$), and assuming "i" multiple discrete electroelastic relaxations, the frequency-dependence of the piezoceramic storage modulus, E' , the loss modulus, E'' , and the loss factor, η , are given by [4]:

$$E'(\omega) = E_r \left[1 + \sum_{i \text{ relaxations}} \Delta_i \frac{(\omega \tau_i)^2}{1 + (\omega \tau_i)^2} \right] \quad (5a)$$

$$E''(\omega) = E_r \sum_{i \text{ relaxations}} \Delta_i \frac{(\omega \tau_i)}{1 + (\omega \tau_i)^2} \quad (5b)$$

$$\eta(\omega) = \frac{E''(\omega)}{E'(\omega)} = \eta_{\max} \frac{2(\omega \bar{\tau})}{1 + (\omega \bar{\tau})^2} \quad (5c)$$

(for a single relaxation)

where τ_{ij} is the i^{th} characteristic relaxation time at constant strain, and $\bar{\tau}$ is the relaxation time:

$$\bar{\tau} = \tau_i (1 + \Delta)^{1/2} \quad (5d)$$

τ_{ij} is given by, for a single piezo segment:

$$\tau_{ij} = R_j C_j^S \quad (5e)$$

where R_j is the shunting resistance and C_j^S the capacitance at constant strain (between the two electrodes on the single segment).

Non-Uniform Strain in Fiber Segments. If the strain within a fiber segment between adjacent electrodes changes sign within the segment, the effective loss factor approach to estimating damping is inappropriate. For illustration, consider the case of a sinusoidal strain distribution. If the wavelength is on the order of the segment length, no voltage appears across the electrodes and, as a result, no damping can occur. However, the effective loss factor approach would predict some damping based on the non-zero strain energy stored in the segment. In practice then, the fiber segment lengths must be considerably shorter than the smallest wavelength of vibration to be damped.

Note that this effect is a result of the fact that with external resistive shunting, piezoelectric damping is not an *intrinsic* property of the material, but an *extrinsic* one, depending on structural length scales.

Shunting Network Topology. As noted in the preceding, the dynamics of the electrical shunting circuit results in frequency-dependent behavior. A circuit comprising a piezoelectric fiber (electrically a capacitor) and a shunt resistor exhibits characteristic exponential relaxation. The time constant of this RC circuit relaxation can be tuned to produce peak damping at a frequency of interest through the suitable selection of the shunt resistance.

For single-segment (monolithic) piezoelectric elements, this selection is fairly straightforward. For more complex circuits with multiple segments experiencing non-uniform strain, deformation of adjacent segments affects the electrical impedance "seen" at the terminals of a given segment. This factor should also be considered in design. Again, ensuring that fiber segment lengths are considerably shorter than the smallest wavelength of interest should minimize this effect.

Damping Performance Analysis: Fiber Effectiveness in Composite Damping

The objectives of this part of the effort were: (1) to develop a theoretical model for prediction of the modal damping of polymer-matrix composite plates with added resistively-shunted piezoelectric fibers; and (2) to use this model to assess the potential effectiveness of such fibers in damping plate vibrations. Previous study of the flexure of composite panels has shown that, in general, the reinforcing fiber (whatever the material) plays a significant role in

damping "fiber-dominated" bending modes, but is less effective in damping "matrix-dominated" twisting modes [7].

A two-part approach was followed in modeling of composite plate damping. The first part involved micromechanical modeling to predict the stiffness and damping properties of a single composite lamina from fiber and matrix properties. The second part involved analytical dynamic modal analysis of a midplane-symmetric laminated composite plate.

Lamina Modeling. Several micromechanical models for calculating lamina elastic properties from constituent properties for two-phase composites are available in the literature. However, in this work, the composite comprises three phases: reinforcing glass fibers, epoxy matrix, and resistively-shunted piezoelectric fibers.

The three-phase composite of interest was treated as a two-phase material by considering the reinforcing fibers and matrix as an effective matrix phase and the piezoelectric fiber as the reinforcing phase. Properties of the effective matrix material were then calculated using micromechanical models valid for isotropic fibers in an isotropic matrix. Because of its two-phase nature, the resulting effective matrix is transversely isotropic.

The piezoelectric fibers were also treated as transversely isotropic, requiring the use of a micromechanical model that is valid for transversely isotropic fibers and matrix to determine lamina properties. A scheme suggested in [8] was used to transform a micromechanical model for transversely isotropic fibers and an isotropic matrix [9] for use with transversely isotropic fibers and matrix.

The lamina elastic models described in the preceding were extended to include damping by representing material damping properties in terms of complex moduli. For harmonic forced vibration of a viscoelastic material, effective dynamic moduli can be determined from expressions for elastic moduli by allowing the elastic moduli to be complex [10]. The real part of a complex modulus is a measure of the stiffness of a material, while the complex part is a measure of damping.

Material Properties. Table 1 summarizes the properties used for the constituent materials in subsequent analysis [11, 12, 13].

Table 1: Constituent Material Properties

	3M S2-glass fiber	Hercules 3501-6 epoxy
Young's modulus, E (GPa)	86	4.3
Shear modulus, G (GPa)	35	2.1
Poisson's ratio, ν	0.22	0.34
Young's loss factor, η_E	0	0.03
Shear loss factor, η_G	0	0.033

Piezoelectric fiber damping was characterized by the (maximum) effective loss factors described in the preceding section. Note that the glass fiber has been assumed lossless, while the epoxy matrix exhibits substantial damping.

Plate Modal Analysis. The modal frequencies and damping of the composite plate were determined from lamina stiffness and damping properties, the laminate stacking sequence and ply orientation, and the plate geometry and boundary conditions. The approach used in this work is similar to that described in [7], which contains additional references to other pertinent work. The following paragraphs provide an overview of the approach.

A higher-order plate theory was used to include shear deformation and rotary inertia effects. Corresponding terms appear in the expressions for plate strain and kinetic energy.

The Rayleigh-Ritz method was used to model the dynamic behavior of the plates under various boundary conditions. The assumed plate mode shapes for both transverse displacements and shear deformation were combinations of beam mode shapes appropriate to the boundary conditions.

Use of these shapes with the strain and kinetic energy expressions and subsequent minimization yielded a complex matrix eigenvalue problem of dimension $4 \times M \times N$. The eigenvalues and eigenvectors were calculated using a standard complex eigenvalue extraction routine.

The resulting complex eigenvalues had the form:

$$\lambda = -\zeta\omega \pm i\omega\sqrt{1-\zeta^2} \quad (6)$$

from which the damped modal vibration frequency, $\omega\sqrt{1-\zeta^2}$, and modal damping ratio, ζ , were readily determined. Note that in the simplest case of a structure made from a single lightly-damped material, the modal damping ratio is approximately half the material loss factor, η , at the corresponding frequency. An effective modal loss factor may thus be defined as twice the modal damping ratio.

Plate Configuration. Table 2 and Fig. 2 illustrate the baseline layup, geometry, and boundary conditions considered in this analysis:

Table 2: Composite Plate Configuration

Layup	Unidirectional
Geometry	10 cm square, 2 mm thick (4-ply)
Boundaries	Cantilevered (CFFF) with fiber direction normal to clamped side

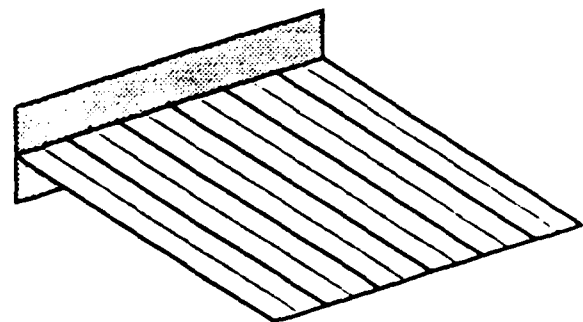


Figure 2: Composite Plate Configuration

Results: Longitudinal Fiber Damping Only. The dependence of plate modal damping on piezoelectric fiber volume fraction was of particular interest in this study. Initially, the only piezoelectric damping of interest was that due to longitudinal stress, with a corresponding loss factor of 0.35 (longitudinal poling).

The epoxy matrix volume fraction was assumed fixed at 0.40 for all cases, while the remaining 0.60 was divided between glass fiber and resistively-shunted piezoelectric fiber. A reasonable upper limit on piezoceramic volume fraction was taken as 0.30. Table 3 summarizes the initial cases considered:

Table 3: Analysis Cases for Longitudinal Fiber Damping

	Piezo Fiber	Glass Fiber	Epoxy Matrix
Case 0	0	0.6	0.4
Case 1	0.1	0.5	0.4
Case 2	0.2	0.4	0.4
Case 3	0.3	0.3	0.4

Table 4 summarizes the calculated *elastic* properties for Cases 0 and 3 (0% and 30% piezo, respectively).

Table 4: Elastic Properties for 0% and 30% Piezo Lamina

	Case 0	Case 3
Longitudinal modulus, E_L (GPa)	53.9	44.0
Transverse modulus, E_T (GPa)	17.4	15.8
Shear modulus, G_{LT} (GPa)	6.9	6.9
Transverse Shear modulus, G_{TT}	6.1	6.1
Poisson's ratio, ν_{LT}	0.26	0.27
Density, ρ (kg/m ³)	1940	3683

Note that the addition of 30% piezoceramic fiber decreases the plate longitudinal stiffness by 18% and increases the density by 90%. Clearly, large increases in damping are needed to justify the use of piezoceramic fibers in a glass/epoxy composite structure.

Figs. 3 and 4 show the dependence of composite plate vibration frequencies and damping on piezoelectric fiber volume fraction. Note that the modal frequencies generally decrease with the addition of piezoceramic fiber, and that the modal loss factors increase dramatically.

The frequencies decrease because the density increases and the stiffness decreases. With the addition of 30% piezoceramic fiber, the largest frequency change is 34%, while the smallest is 30%. By sensitivity, the modes are ordered 1, 4, 5, 2, 6, 3. Mode 1 is a "fiber-dominated" bending mode, while Mode 3 is a "matrix-dominated" twisting mode. The behavior of the other modes is between these extremes.

Table 5 summarizes the effective modal loss factors for Cases 0 and 3 (0% and 30% piezo, respectively), along with the percent changes in damping. With the addition of 30% piezoceramic fiber, Mode 1 damping increases by a factor of 50 to a level of over 12%. Mode 3 is affected the least, but nevertheless increases by 42% to a level of 3.6%. This result

agrees with previous findings regarding the role of the fiber in composite damping. Note that for non-unidirectional fiber arrangements and for shell-type structures that carry in-plane loads, fiber damping would play an important role in most vibration modes.

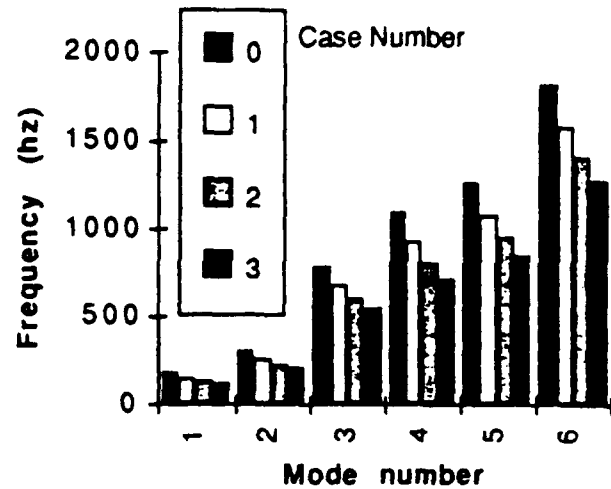


Figure 3: Modal Vibration Frequencies versus Mode Number and Piezo Volume Fraction Case

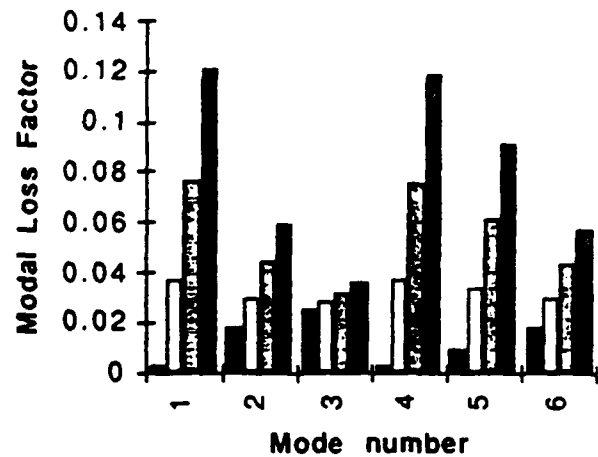


Figure 4: Modal Loss Factors versus Mode Number and Piezo Volume Fraction Case

Table 5: Modal Loss Factors for 0% and 30% Piezo Lamina

	Case 0	Case 3	% increase from Case 0
Mode 1	0.0024	0.1205	4920%
Mode 2	0.0173	0.0589	240%
Mode 3	0.00251	0.00357	42%
Mode 4	0.0027	0.1183	4280%
Mode 5	0.0091	0.0908	987%
Mode 6	0.0180	0.0569	216%

Results: Transverse Fiber Damping Added. The preceding analysis is based on the assumption that only longitudinal fiber stress contributes to modal damping. Because each fiber segment has only a single pair of electrodes, transverse deformation may increase or decrease the potential difference between them, depending on the sign of the transverse stress. This will change the apparent damping, increasing or decreasing it according to the relative signs of the stresses.

An upper bound for the effects of a more complex stress state involving transverse deformation may be established by assuming that the effective loss factors for longitudinal and transverse stress act additively. A lower bound may be similarly obtained. For longitudinal fiber segment poling, the longitudinal loss factor was taken to be 0.35, while the transverse loss factor was taken to be 0.06.

Table 6 summarizes the effective modal loss factors and changes for Cases 3 and 3T. Both cases have 30% piezo fiber, but Case 1 assumes longitudinal piezoceramic damping only, while Case 3T also includes additive transverse damping.

Table 6: Modal Loss Factors with Added Transverse Fiber Damping

	Case 3 (L only)	Case 3T (L and T)	% change 3 to 3T	% change referred to Case 0
Mode 1	0.1205	0.1233	2.3%	117%
Mode 2	0.0589	0.0601	2.0%	7%
Mode 3	0.0357	0.0363	1.7%	2%
Mode 4	0.1183	0.1211	2.4%	104%
Mode 5	0.0908	0.0927	2.1%	21%
Mode 6	0.0569	0.0580	1.9%	6%

Additional changes on the order of 2% are seen with the inclusion of the transverse loss factor. The dominant contributor to increased composite damping is clearly the longitudinal loss factor. This is a result of the higher loss factor in combination with greater participation in modal strain energy.

A more detailed analysis of the effects of complex stress states would likely require a numerical coupled-field approach, similar perhaps to that described in [14].

Experimental Results. No true resistively-shunted piezoelectric fiber composites have yet been constructed, although this analysis indicates their potential to exhibit extraordinarily high levels of damping for structural materials. Experimental results validating the general concept of shunted piezoelectric damping using monolithic elements have been reported elsewhere in the literature [1-4].

Piezoelectric Fiber Fabrication

Researchers have recently fabricated 50 micron diameter PZT fibers ($\text{PbZr}_{0.52}\text{Ti}_{0.48}\text{O}_3$) from viscous spinnable solutions prepared by sol-gel processing of alkoxide precursors. Following heat treatment, these fibers were found

to consist of fully dense submicron grains, and to exhibit dielectric constants of 800. This progress is discussed in detail in [15].

The additional critical aspects of fiber electroding, poling, and integral shunting are currently under investigation. Various alternative approaches are being considered for each.

Summary and Conclusions

In summary, there is a need for structural materials with enhanced intrinsic vibration damping capability. While researchers have recently demonstrated the use of resistively-shunted piezoelectrics to increase structural damping, these efforts used elements with dimensions of the same order as structural elements. The extension of this emerging research area to composite materials with tailorable frequency-dependent damping, along with corresponding design and analysis tools shows promise. With longitudinally-poled fibers, peak modal loss factors of 12% are theoretically attainable in a PZT/S-glass/epoxy composite, even at low (30%) piezoceramic fiber volume fractions. Successful pursuit of this avenue of development would mark a significant advance in the technology of engineered structural materials.

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